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AFOSR MURI
Search for New and Better High Temperature Superconductors
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Abstract

This program was focused on an integrated search for new superconductors in material systems with perceived potential for higher temperature superconductivity. The results have provided a greatly improved understanding of the high-T_c bismuthate superconductors and their potential for even higher T_c's , along with related mixed-valent materials. New pnictide and related materials have also been discovered and/or studied that go beyond the canonical Fe-based pnictides in composition and/or structure. The results provide new insights and opportunities for higher T_c superconductivity in this class of materials. Careful examination of two select material systems for which very high-T_c trace superconducting anomalies have been reported has demonstrated interesting new physical effects. The relevance of these effects to possible high-T_c anomalies is unclear, however. A new scanning probe (a Scanning Tunneling Potentiometer) has been developed that has great potential for studying transport (especially superconducting) in thin films on the nanometer scale.

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1) Introduction

This program was focused on an integrated search for new superconductors in material systems with perceived potential for higher temperature superconductivity. We also examined select materials for which credible high-T_c superconducting anomalies have been reported, and we developed a new tool (scanning tunneling potentiometry) for nanometer-scale transport characterization of thin films. In addition, we actively addressed the questions: (1) Can modern materials theory play a useful role (along with chemical intuition) in the search for new superconductors? And (2) Are there physical limitations to the superconducting transition temperature independent of the mechanism?

The team consisted of Malcolm Beasley and Ian Fisher at Stanford, Robert Cava at Princeton, Gabriel Kotliar at Rutgers and Emilia Morosan at Rice. Our expertise included bulk synthesis, thin-film synthesis, materials theory and novel scanning probes. Both physics and solid-state chemistry perspectives were represented.

In the next section we briefly summarize our results, trying to put them in useful context. There is also an Appendix in which we list all the publications resulting from the MURI. This appendix also serves as the endnotes indicated in the text of the Summary of Results.

2) Summary of Results

2.1) *Bismuthate Superconductors – The archetypical mixed-valent superconductor*.

As is well known, a very large effort has been focused on understanding the superconductivity in the high-T_c cuprate and Fe-based (or pnictide) superconductors, for which in both cases electron correlations are thought to play an important role. Less celebrated, but similarly interesting, are the bismuthate superconductors. They have moderately high T_c's and have defied quantitative understanding for over 20 years. Their full potential for higher T_c's has been heretofore unknown. It is this class of superconductors and related materials that have been a major focus of this MURI.

So-called mixed-valent materials are those for which a valence-skipping element is present and disproportionates to avoid the unfavorable valence state (e.g., Bi^{4+} -> Bi^{3+} + Bi^{5+}, as in the archetypical mixed-valent bismuthate $BaBiO_3$). The excess

positive (and negative) charge on the two sites is screened by motion of the neighboring oxygen atoms (breathing mode) around the charge-disproportionated valence-skipping element. In the end, this charge disproportionation leads to a charge density wave (CD-CDW) and an associated energy gap in the electronic structure of the material. Note that a CD-CDW is not due to Fermi surface nesting; it is distinct form of CDW. Note also that the charge disproportionation involves the physics of both valence skipping and motion of the lattice.

Doping $BaBiO_3$ (BBO) on the A or B sites (with K or Pb, respectively, and known as BKBO and BPBO) leads to a region of superconductivity adjacent to the CD-CDW phase. In the case of BKBO the maximum T_c is ~ 30K. Traditional LDA electronic structure calculations predict $BaBiO_3$ to be a simple half-filled-band metal. Similarly, the most advanced LDA-type calculations yield an electron-phonon interaction parameter λ that is too low to account for the high T_c's exhibited in the doped bismuthates.

2.1.1 *Importance of correlation effects in the bismuthates*

A key advance in the understanding of the bismuthates came in the work of Franccini and co-workers, who showed that DFT calculations using the HSE hybrid density functional could account quantitatively for the CD-CDW found in $BaBiO_3$. This computationally more intensive HSE functional corrects the tendency of LDA calculations to overestimate screening. The HSE functional (and equivalent approaches) accounts for the non-local dynamic correlations present in the bismuthates. Simply put, the bismuthates are correlated materials. Kotliar has dubbed them the "other" class of correlated materials, distinct from the cuprates and the pnictide superconductors.

Building on the work of Franccini, the Kotliar group showed that these correlations lead to dynamically-enhanced λ's that can account for the high T_c's found in the bismuthate superconductors [25]. They also calculated the optical conductivity (using dynamical mean-field theory) and found good agreement with experiment [23]. Importantly, their calculations demonstrated that the correlations shift oscillator strength in the real part of the conductivity $\sigma_1(\omega)$ to higher energies where it cannot contribute to the superfluid density in the superconducting state – an unfortunate consequence of correlation also found in the cuprates and pnictides. Based on this new understanding, the Kotliar group then proposed a rational design approach for mixed-valent high-T_c superconductors [24]. Finally, building on their work on the bismuthates, the Kotliar group went on to show that their new formalism could also account for the previously unexplained relatively high temperature superconductivity found in electron-doped MNX (M = Ti, Zr and Hf; X = Cl, Br and I) [25]. It will be interesting to see how large this "other" class of correlated superconductors turns out to be.

2.1.2 *The importance of disorder (Anderson localization) on the T_c's of the bismuthate superconductors*

In addition to correlation, we have also found that disorder (Anderson localization) can have a strong affect on the T_c's of the bismuthate superconductors [8]. More specifically, the Stanford Group (Beasley, Fisher and their students) showed using point contact tunneling spectroscopy that there was a distinct square-root cusp around zero bias in the tunneling density of states of single crystals of BPBO . Such behavior is characteristic of enhanced Coulomb interactions due to localization. These enhanced Coulomb interactions lead to an increase in the renormalized Coulomb interaction parameter μ^* that enters the famous expression for the superconducting transition temperature $T_c = \Omega_0 \exp(1/(\lambda - \mu^*)$. An increased μ^* therefore decreases T_c. Using the most highly developed theory of these effects, the Stanford group was able to show that the disorder-free maximum T_c in BPBO is a factor of two higher than that observed, whereas in BKBO the enhancement is small, in keeping with the lower resistivities (and hence disorder) observed in this case.

In addition, the Stanford Group observed a linear decrease in the zero-temperature conductivity σ_0 as a function of Bi doping x in $BaPb_{1-x}Bi_xO$ (BPBO). Such behavior is expected in the theory of localization and the extrapolation of this data to zero conductivity showed that there is a disorder-induced metal/insulator transition at $x \sim 0.26$, which clearly occurs below the composition at which the CD-CDW state is reached ($x \sim 0.35$). Unfortunately, there is presently no theory that treats CD-CDW formation in the presence of Anderson localization. We hope that our work will stimulate such theoretical effort.

This new methodology for determining the disorder-free T_c in s-wave superconductors with large microscopic disorder should have a much larger range of application than just to the bismuthates. As a start in this direction, the Beasley group applied the methodology to K-doped WO_3 and found a disorder free $T_c \sim 30K$, a remarkable factor of 6 greater than the observed T_c [9]. As discussed later, this adds fuel to the interest in the reports of very high-T_c anomalies on (or near) the surface of WO_3 when covered with alkali metals.

Also, in collaboration with the Morosan group, the Beasley group has looked at Cu-intercalated $TiSe_2$, which exhibits superconductivity and a conventional CDW. In this case they found the presence of enhanced Coulomb effects but the affect on T_C was small [1].

2.1.3 *Other new physical phenomena*

The existence of correlation effects and an Anderson metal/insulator transition in BBPO were not the only new physical effects discovered by the MURI. Using x-ray and neutron scattering, the Cava group showed that the microstructure of BPBO in the region of the superconducting dome was dimorphic (i.e., exhibiting the same composition but different local structures) [54]. Both tetragonal and orthorhombic phases were evident. Also, the size of the Meissner fraction observed correlated with the amount of the tetragonal phase as the doping x varied across the superconducting dome, suggesting that superconductivity was associated with this phase.

Building on the work of the Cava, the Fisher group in collaboration with Kramer at Iowa State generated and analyzed an extensive series of HRTEM images for samples spanning the dimorphic region [5]. They use Fourier filtering to provide contrast between the orthorhombic and tetragonal regions. Using spatial correlation functions derived from these images, they established that the nanostructure present was stripy in character and oriented along definite crystallographic directions. They also were able to characterize the various length scales observed in the nanostructure. The results show that the superconducting coherence length is larger than all the characteristic length scales at low doping but decreases to be on the smaller scale of the nanostructure at optimal T_c.

This result provides a means to understand the existence and character of the superconducting dome in BPBO. At low compositions the size of the Cooper pairs is larger than the nanostructure and the disorder in the material affects T_c through the enhanced Coulomb interactions discussed above. Beyond the peak, the superconductivity is granular in nature, and T_c is suppressed by phase fluctuations. While still qualitative in many respects, an understanding of the existence and character of the superconducting dome observed in BPBO now seems to be emerging. We note that in the case of BKBO, where disorder is much smaller, there is no superconducting dome. The superconducting region ends abruptly at the boundary of the CD-CDW region of the phase diagram.

Another striking phenomenon found in BPBO is a superconductor/insulator quantum phase transition as a function of magnetic field that occurs most clearly in the vicinity of the maximum T_c in the superconducting dome [34]. Very good scaling of the transition as a function of T and H is observed with critical exponents typical of a two-dimensional superconductor. This is very surprising in that BPBO is a three-dimensional material. While the phenomenon is undeniable, it origins and character are not understood at the present time, as does any possible role of the stripy nanostructure in the region of the superconducting dome as a function of composition discussed above.

2.2 New Materials Directions I – Other mixed-valent materials

The work described above has spawned new material directions. For example, using the new theoretical framework they developed, the Kotliar group has predicted that La-doped $BaPbO_3$ (LBPO) should be a 15 to 20K superconductor. Undoped $BaPbO_3$ is a band metal and not superconducting. Doping with La would be expected to add electrons to the material and move the Fermi level to higher energies. However, that is not the important point; there is a new physical insight here. The Kotliar group found theoretically that the strong el-ph interaction in BPO occurs for electrons in the band above the original conduction band, hence the need for large electron doping. This insight also provides and explanation why $BaPbO_3$ does not charge disproportionate, even though Pb is a well-known valence-skipping element.

Motivated by this calculation, the Beasley group in collaboration with the Schlom/Shen group at Cornell, synthesized thin films of LBPO and studied their properties by ARPES, transport and point contact tunneling spectroscopy. These groups found that the observed Fermi level is in the higher conduction band as predicted but that the films were insulating. This seeming contradiction was resolved by more extensive transport and point contact tunneling data that showed the now familiar signs of disorder-induced electron localization. The next step would be to try charge-transfer doping using the ionic liquid approach in order to avoid disorder.

We also note, that the zero-bias cusp in the density of states due to the enhanced Coulomb interactions was observed in both the point contact spectroscopy and in angular averaged photoemission. The two measurements were in quantitative agreement in the energy range of the cusp but deviated at higher energies for reasons that are not clear.

In another example, the Cava group carried out a computational survey using conventional LDA calculations and found that the electronic structure of $RbTlCl_3$ is very close to that of $BaBiO_3$, as shown by the similar Fermi surfaces (in the LDA) of these two materials. The next step is to synthesize and dope the material. Cava is a card carrying new materials experimentalist, and his willingness (actually long standing) to use theory in the pursuit of new superconductors will surely catch the attention of the more traditional new superconducting materials researchers.

Motivated by the recent work on the bismuthates, and based on knowledge that the Au in CsAuBr3 and CsAuI3 exhibit mixed valence, the Fisher group attempted chemical doping these materials. They found that they were very hard to dope and could not be made metallic by chemical doping. On the other hand, in collaborations with others, they were able to achieve a better understanding of the pressure tuned CD-CDW to metal transition observed in these materials [19]. Their work stimulated the Kotliar Group to theoretically investigate the el-ph coupling in these materials. Their work suggests that there is strong el-ph coupling associated with the breathing mode if the Fermi energy could be increased enough through electron doping.

Finally, the Kotliar group examined theoretically $CsTlF_3$ and $CsTlCl_3$ [24]. Since Tl is known to be a valence-skipping element, these materials are good candidates for charge disproportionation. Indeed, the Kotliar group calculations confirm this and predict that superconductivity with $T_c > 30$ K, if the materials could be doped.

Based on these predictions, Kotliar collaborated with his colleague Martha Greenblatt in the Chemistry Department at Rutgers to test the predictions. They studied hole doping of $CsTlCl_3$ by substitution of Tl with Hg, in an attempt to metalize the $CsTlCl_3$ compound and hopefully induce superconductivity at low temperatures. Kotliar's ab initio supercell calculations uncovered a common theme of the structures of the doped $Cs(Tl,Hg)Cl_3$ compounds: there are equal amount of Tl3+ and Tl1+ ions with breathing-distorted Cl- ions surrounding them, while the Cl-

ions around the Hg2+ ions form second order Jahn-Teller distorted octahedrons, similar to the Jahn-Teller distortion in the parent compound $CsAuCl_3$. These distortions maintain the insulating nature of the parent compound in the whole doping range of $Cs(Tl,Hg)Cl_3$. These theoretical results are fully consistent with experimental measurement and provide important insights into why Hg doping does not metalize the $CsTlCl_3$ parent compound. Further work is needed to metalize the parent by substitutions of other components.

2.3 New Materials Directions II -- Pnictide superconductors and related materials

The Rice group focused its work on the search for novel layered transition metal pnictides and understanding competing ground states at the itinerant-to-local moment crossover where new phenomenon are most likely to be found. This work stands in contrast to most other groups, which focus mainly on pnictides containing Fe. As a result of this work, two series of new compounds with stoichiometry $R_3T_4As_4O_2$ have been discovered and characterized (R = rare earth, T = Cu or Ni). Varying the R ion led to the observation of diverse physical properties including superconductivity for R = La and T = Ni, ferromagnetic or antiferromagnetic order for R = Ce, Pr and Sm, and spin glass behavior in $Nd_3Ni_4As_4O_2$. In the Ce members of the series, $Ce_3Cu_4As_4O_2$, remarkable magneto-transport behavior is unveiled [6], pointing to a quantum critical point induced by a magnetic field, accompanied by non-Fermi liquid behavior, similar to that observed in heavy fermion intermetallics.

In another study, motivated by the theoretical suggestion by the Kotliar group that hypothetical $BaFePn_2$ compounds would have metallic blocking layers sandwiched between the FePn tetragonal layers, the Rice group found the experimental realization in a related transition-metal pnictide, $SrMnBi_2$ [46]. In $SrMnBi_2$, the Mn-Bi tetrahedral are closer to the ideal tetrahedron (and likely favorable for high T_c superconductivity) than the better known Fe pnictide superconductors, as well as the theoretical compounds $BaFePn_2$. The ordering temperature is close to 300 K, indicating the possibility of commensurately high T_c once the magnetic state can be suppressed.

In another new materials direction, the Rice group also studied checkerboard Mott insulators: $(SrF)_2Fe_2OS_2$, and $(LaO)_2Fe_2OS_2$ [7, 26]. These are compounds with a layered structure comprised of A^{2+} layers (A = SrF or LaO) separated by T_2OX_2 layers (T = magnetic transition metal, X = S or Se) where T ions form a square lattice, with O ions at the center, and X ions above and below alternating T plaquettes. These materials are Mott insulators, promising avenues for searching for new high temperature superconductors with doping or pressure. So far we have synthesized the pure compounds with T = Fe and X = S in polycrystalline form. Doping experiments have so far indicated that the magnetism is very robust such that both the ordered state as well as the insulating behavior can hardly be altered.

They also discovered a new Fe pnictide compound with tetragonal FeAs stripes, CaFe4As3 [60], with SDW AFM order around 90 K [44], comparable to the parent compounds for the layered Fe superconductors. Doping and pressure [48]

consistently show complex magnetism (up to three magnetic phases below T_N) but with a structural distortion taking place before the magnetic state can be suppressed in favor of superconductivity.

Finally, P doping into Co_2As was used as a way to possibly induce a local-to-itinerant moment crossover, where superconductivity is often found. Instead the Rice group found that the isoelectronic doping of Co_2As induced two structural modifications, resulting in a ferromagnetic state at intermediate P compositions [4].

The Princeton group also focused on novel pnictides. One of the directions explored arose in an unusual way. During the course of our MURI program, a researcher in Japan who was not knowledgeable in the techniques or concepts of solid-state chemistry, reported the existence of new arsenide superconductors in the Ca-Fe-Pt-As chemical system with T_c*'s* up to 38 K. Although layered -Ca-(Fe, Pt, As)-Ca-(Fe_2As_2)- stacking was suggested for the structures of the superconductors, neither the crystal structures nor the detailed physical properties were characterized. One of the major efforts by the Princeton group as part of this MURI was to determine the crystal structures and physical properties of two superconductors in this chemical system, $Ca_{10}(Pt_3As_8)(Fe_2As_2)_5$ (the \10-3-8 phase") and $Ca_{10}(Pt_4As_8)(Fe_2As_2)_5$ (the \10-4-8 phase").

This was a decidedly difficult endeavor. Still, by using the high temperature flux growth method, the Cava group was able to grow mm-sized 10-3-8 phase single crystals from a CaAs flux. The 10-4-8 phases proved to be even more difficult and only smaller crystals were obtained. The crystal structures, which are complex, were then determined from the single crystal X-ray diffraction experiments. These new materials raised the chemical complexity of the superconducting pnictides to a new level, making them particularly interesting from a solid-state chemistry perspective. For example, based on -Ca-(Pt_nAs_8)-Ca-Fe_2As_2- layer stacking, the 10-3-8 phase has triclinic symmetry, which is very rare for superconductors, while the 10-4-8 phase has tetragonal symmetry. (The triclinic symmetry was later shown by ARPES to have relatively little effect on the electronic structure of the layers.) The FeAs layer is made of edge-sharing $FeAs_4$ tetrahedra, which are the same key structural ingredient that is present in all Fe pnictide superconductors. The intermediary Pt_nAs_8 layer is quite different, however. It is a derivative of the commonly found skutterudite structure, which consists of a square lattice of corner-sharing $PtAs_4$ squares whose orientation is governed by the formation of intraplanar As-As dimers.

The Princeton group found that for the triclinic 10-3-8 phase, the superconducting T_c can be tuned from 0 to 11 K through substitution of 6% to 13% of Pt on the Fe sites in the FeAs layers. The Pt substitution dopes the layers with electrons. Although the ground state of the undoped 10-3-8 phase remains unclear even now, the Pt doping study provided evidence for similarities between this new superconducting family and the frequently studied 1111 (such as $LaFeAs(O_{1-x}F_x)$) and 122 (such as $Ba(Fe_{1-x}Co_x)_2As_2$) families of arsenide superconductors. For the tetragonal 10-4-8 phase, 13% Pt on the Fe sites stabilizes superconductivity at a much higher temperature, 25 K, which is more than twice the highest T_c observed in the 10-3-8 phase. Of particular interest to the Cava group was the fact that these two chemically and structurally similar compounds with

significantly different T_c*'s* provided a particularly interesting platform for studying superconductivity in the pnictides.

From the chemical point of view, the d^8 Pt^{2+} and the formation of As-As dimers (As_2^{4-}) in the Pt_nAs_8 layers leads to an intermediary layer of $[Pt_3As_8]^{10-}$ for the 10-3-8 phase and $[Pt_4As_8]^{8-}$ for the 10-4-8 phase. Thus in its undoped form, the 10-3-8 phase is a valence satisfied compound through the Zintl concept - the $[Pt_3As_8]^{10-}$ layer is perfectly charge balanced by the $[Ca10]^{20+}$ and $[Fe_{10}As_{10}]^{10-}$ layers, leading to semiconducting character for the Pt_3As_8 intermediary layer (i.e., it will not contribute density of states at E_f) . This is then expected to lead to weak FeAs interlayer coupling through the Pt_3As_8 intermediary layer. The 10-4-8 phase on the other hand has one more Pt atom in the Pt_4As_8 intermediary layer, which therefore exceeds its valence satisfaction requirements, indicating that this layer is likely to have states at E_f and is therefore metallic in character - leading to stronger FeAs interlayer coupling.

From the structural point of view, these new superconductors do not follow the usual empirical structural rule between T_c and the As-Fe-As bond angle or *Pn* height (the distance between the adjacent Fe and As layer) that is often invoked to explain the origin of the different T_c*'s* in the arsenide superconductors, rather, significant structural differences that can affect the interlayer coupling are present. In the 10-3-8 phase, there is only one such Pt_3As_8 to FeAs interlayer interaction per unit cell, and it is irregular due to significant disorder in the crystal structure. This not only leads to weaker interlayer coupling from the structural perspective, but also helps to electronically isolate the Pt_3As_8 intermediary layer from the FeAs layers, making Pt_3As_8 layer more electronically blocking and reinforcing its semiconducting nature. In the 10-4-8 phase, on the other hand, due to two rather than one channel interlayer interactions, and their structurally ordered character, stronger interlayer coupling is realized and the metallic nature of the Pt_4As_8 layer is reinforced.

Given the very similar crystal structures and chemistry of these two superconducting Ca-Pt- Fe-As compounds, comparison between the low *Tc* 10-3-8 phase and the high T_c 10-4-8 phase leads to the understanding that the differences caused by stronger FeAs interlayer coupling through the intermediary Pt*n*As8 layer in the 10-4-8 phase, due to its two-channel interlayer interactions and its metallic nature, is what gives rise to its higher T_c. Thus the analysis of these phases shows that stronger interlayer coupling is a important factor in enhancing T_c in the superconducting Fe pnictides. The relatively high T_c of the 10-4-8 phase made it of significant interest in the materials physics community, and many different collaborators worked to elucidate its properties, with materials synthesized in our laboratory, during the course of the MURI program. Of particular interest to the Princeton group was the research performed by an experimental group at the IOP in China that showed that there was a correspondence between the superconductivity induced by pressure and the superconductivity induced by chemical doping in the 10-3-8 phase. This is a surprising result, reminiscent of the behavior of doped Mott insulators such as V_2O_3, which has not yet been followed up much in the community.

2.4 *Trace High-T_c Anomalies*

There are many reports of trace superconductivity at very high temperatures, but two stand out as the most credible. The first is at the interface of Cu metal deposited on the antiferromagnetic Mott (charge transfer) insulator CuO and then subjected to various (uncontrolled) heat treatments by application of very high current densities through the Cu layer. Our approach was not to try to replicate the heat-treatment results but rather to study carefully the as-deposited Cu/CuO interface to see if there were any signs of new physics that might suggest the potential for high-T_c superconductivity. While no superconductivity was detected, we did indeed find new physics. By contrasting the low temperature transport properties of Cu deposited on CuO with that deposited on the conventional insulator MgO, we found a significant difference in the weak localization regime well known in Cu due to the presence of trace impurities. Specifically, we found that the suppression of localization due to Fe impurities in the Cu/CuO case was much less than for the Cu/MgO case [2]. We concluded that the apparent weakening of magnetic scatter in the Cu/CuO case was due to antiferromagnetic correlations induced into the Cu via a spin-channel proximity effect with the antiferromagnetic Mott insulator CuO. Thus, although no superconductivity was observed, we did find a wholly new kind of proximity effect. What role these antiferromagnetic correlations might play in any high-T_c anomalies is an open question.

The second case of interest was the persistent reports of very high temperature superconductive signatures on the surface of WO_3 crystals treated by alkali metals. Our approach was to search for these phenomena on thin films of WO_3 where more sophisticated in situ physical and in situ surface materials characterization could be carried out. This turned out to be a major challenge in thin film growth. The films showed highly variable properties until a process was discovered empirically that led to control of the oxygen deficiency. Once this was achieved, we carried out systematic studies of both doped and undoped WO_3. It was in such studies that we found that the disorder free T_c was higher by a factor of 6 than the actual T_c, as detailed in Section 2.1.2 above [9].

2.5 *Fundamental physical limits to T_c*

One of the motivating factors in this MURI was an appreciation that both reduced dimensionality and low superconducting pair density are not favorable for very high-T_c superconductivity. The physical reason is that phase fluctuations of the superconducting pair wave function are enhanced under these conditions and therefore can limit the transition temperature, even if pairing occurs at higher temperatures. This was among the reasons to look at potentially high-T_c materials that were three dimensional, such as the bismuthates.

During this MURI program, the conflict between strong interactions and reduced pair density has taken sharper focus. We now know from the cuprates, pnictides and bismuthates that electron correlations play a significant role in reducing the pair density. In all these cases, oscillator strength in the optical conductivity is

shifted to higher energies where it cannot participate in the zero-energy delta function in $\sigma_1(\omega)$, the weight of which determines the superfluid density.

The news is not all bad, however. Stimulated by our earlier arguments, Kivelson and coworkers showed theoretically via a model calculation that this problem can be avoided by means of a proximity route to higher T_c in which coherent tunneling between a high-density metal layer and an underlying negative-U material (as a model system) can yield simultaneously high T_c and high carrier density. The key point is that the superconductivity is induced in the normal metal, even if the negative U material is insulating. The Hamiltonian used in these calculations is isomorphic to a material with two bands – one conducting and the other containing the negative U centers. The important lesson here is that the problem of the reduction of the superfluid density with the shift of oscillator strength to high energies is a property of one-band materials. This insight gives important guidance in assessing materials for their potential to exhibit high temperature superconductivity.

2.6 Nanometer-Scale Scanning Tunneling Potentiometry

Interesting new superconductors are often inhomogeneous, due to materials science issues or perhaps even intrinsically. For some time, we have been working to develop a tool that can directly study the electron conductivity in such inhomogeneities through electrical transport measurements. Also, this approach should be ideal for detecting trace very high-T_c regions in an inhomogeneous thin film for which trace high-T_c superconductivity appears to be present. Our approach was scanning tunneling potentiometry -- a four point measurement in which one of the contacts is an STM, which is used alternately point by point to generate a simultaneous image of the topography and the potential of a material.

It turned out that his was a far more difficult technical problem than we originally imagined. Happily, we have finally succeeded, although time did not permit study of superconductors. The details and protocol for use or our instrument have now been documented and the capability demonstrated on so-called epitaxial graphene as a model system [17]. This work demonstrates the remarkable capabilities or scanning tunneling potentiometry to make transport measurements on a nanometer scale and, incidentally, to show how complicated transport really is in epitaxial graphene when you look at such small length scales.

Appendix A

List of Publications Under The MURI

2015

1) Katherine Luna, Phillip M. Wu, Justin S. Chen, Emilia Morosan, and Malcolm R. Beasley, *"Point-contact tunneling spectroscopy measurement of CuxTiSe2: Disorder-enhanced Coulomb effects",* Phys. Rev. B **91**, 094509 (2015).

2. Wu, Phillip M.; Ishii, Satoshi; Tanabe, Kenji; et al., *"Synthesis and ionic liquid gating of hexagonal WO3 thin films",* Appl. Phys. Lett. **106**, 042602 (2015).

3. M. Retuerto, Z. P. Yin, T. J. Emge, P. W. Stephens, M. R. Li, T. Sarkar, M. C. Croft, A. Ignatov, Z. Yuan, S. J. Zhang, C. Q. Jin, R. P. Sena, J. Hadermann, G. Kotliar, M. Greenblatt, *"Hole doping and structural transformation in CsTl1-xHgxCl3",* Inorg. Chem. 54, 1066-1075 (2015).

4. Chih-Wei Chen, Andriy H. Nevidomskyy and E. Morosan, *"Enhanced ferromagnetism induced by structural phase transitions in $Co_2As_{1-x}P_x$",* Phys. Rev. B (under review).

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Program Manager

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Abstract

This is the final report on our MURI program focused on an integrated search for new superconductors in material systems with perceived potential for higher temperature superconductivity. The major results are summarized. The program has provided a greatly improved understanding of the high-Tc bismuthate superconductors and their potential for even higher Tc's, along with related mixed-valent materials. New pnictide and related materials have been discovered and/or studied that go beyond the canonical Fe-based pnictides in composition and/or structure. The results provide new insights and opportunities for higher Tc superconductivity in this class of materials. Careful examination of two select material systems for which very high-Tc trace superconducting anomalies have been reported has demonstrated interesting new physical effects. The relevance of these effects to possible high-Tc anomalies is unclear, however. A new scanning probe (a Scanning Tunneling Potentiometer) has been developed that has great potential for studying transport (especially superconducting) in thin films on the nanometer scale. Also, the program demonstrated that modern materials theory is an important element in the new materials discovery process, even for materials in which the

conventional approaches breakdown and electron correlations play an important role.

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Changes in research objectives (if any):

Change in AFOSR Program Manager, if any:

Extensions granted or milestones slipped, if any:

Modification 9 extended the project from 60 to 65 months.

AFOSR LRIR Number

LRIR Title

Reporting Period

Laboratory Task Manager

Program Officer

Research Objectives

Technical Summary

Funding Summary by Cost Category (by FY, $K)

	Starting FY	FY+1	FY+2
Salary			
Equipment/Facilities			
Supplies			
Total			

Report Document

Report Document - Text Analysis

Report Document - Text Analysis

Appendix Documents

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